Self-Assembled Magnetic Dots, Antidots, Dot Chains, and Stripes: Epitaxial Co on Ru(0001) (INVITED)

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Lateral magnetic nanostructures have been grown via molecular beam epitaxy in ultrahigh vacuum and characterized *ex-situ* with atomic force and magnetic force microscopy. We observed that epitaxial growth of Co onto Ru(0001) at elevated temperature results in three-dimensional Co islands (dots) or a flat Co film network with deep holes (antidots) in truncated pyramidal shapes. The lateral size of these dots/antidots, in the order of 100 nm, tends to be uniform at each given coverage. We attribute the growth mode mainly to strain relaxation of Co epitaxy on Ru, which has a 8% lattice mismatch. In addition, we have explored the placement of these dots on a grooved Ru(0001) surface. The dots automatically align into linear chains along the asymmetric grooves to form either dot chains or continuous stripes, which would open new opportunities in creating either ordered magnetic arrays or arbitrary arrangements.

I. Introduction

The fabrication of lateral magnetic nanostructures via self-assembly is attracting increasing interests for both basic research and technology development.[1,2] The physical size of a magnetic system affects its magnetic properties by altering its dimensionality, structure, surface/interface electronic structure, quantum size effects and transport, and domain/domain wall structure and motion. In addition, patterned media[3,4] with individual magnetic dots has been actively explored as an alternative route for ultrahigh density information storage, since having one bit per dot promises to improve signal-to-noise ratio significantly. Self-assembly not only offers an intrinsically simpler, faster, and less costly alternative route to lithography, but also promises thermodynamically stable structures even beyond the lithographic limits. Self-assembled quantum dots have been realized in semiconductor systems,[5] such as Ge/Si, GeSi/Si, and InAs/GaAs, where a strain-driven Stranski-Krastanov (SK) growth mode results in dots of narrow size distributions. Metal self-assembly has been observed in a few systems with one or several monolayer (ML) thick 2D structures utilizing unique surface phenomena such as step decoration,[6,7,8,9] surface strain[10,11] and reconstructed surfaces as templates.[12,13] In this paper we review our recent work on a new metal-on-metal self-assembly mode, where magnetic lateral structures of 70-600 nm wide and 1-7 nm thick form mainly due to strain relaxation in epitaxy.[14,15] For Co grown on flat Ru(0001) at elevated temperature, we observed both 3D islands (dots) or holes in film networks (antidots in smooth films) with well-defined shape, surprisingly smooth surfaces, and relatively narrow size distribution.[14]

A general challenge in self-assembly is associated with placement and alignment of the structures at predetermined locations, a task that is routine for lithographic

fabrication. For example, magnetic patterned media[3,4] consisting of a magnetic dot array would require dot alignment along tracks. It would even be more demanding to produce complex spintronic devices[16] by means of self-assembly. It is, therefore, critical to develop methods to align self-assembled magnetic nanostructures. We have aligned self-assembled magnetic dots along substrate grooves that form due to residual scratches from the mechanical surface polishing used in preparing the substrate.[15] The results offer the promise that magnetic dot arrays or arbitrary arrangements could be fabricated by self-assembly with the assistance of lithographic substrate patterning.

II. Experiment

The experimental details have been reported elsewhere.[14,15] The Co samples with wedge-like thickness gradient of 0 – 420 nm were grown with molecular beam epitaxy (MBE) on smooth and grooved Ru(0001) crystal substrates at 350°C in a ultrahigh vacuum (UHV) system with base pressure of $6x10^{-11}$ Torr. The grooves result from the residual polishing lines or unexpected step bunching, which are straight or slightly curving along an arbitrary direction with a period of ~ 0.5 – 2 μm and height of ~ 4 – 16 nm. The surface was cleaned *in-situ* by cycles of O_2-annealing at 1300 – 1500 K and flashing at 1500 –1600 K.[17] The resultant Ru surface, and the subsequent Co surfaces, are free of O and C contamination as determined with Auger electron spectroscopy. Sharp hexagonal low-energy electron diffraction (LEED) patterns were obtained both on the clean substrate and along the clean Co wedge with no apparent broadening. The Co samples were then covered with a thin layer of Au (< 1 nm) at room temperature to reduce the oxidation of Co in air. The morphology and magnetic imaging of the wedges

were taken *ex situ* with atomic force microscope (AFM) and magnetic force microscope (MFM).

III. Results and Discussions

Figure 1 shows a variety of self-assembled Co lateral structures, i.e., (a) dots, (b) antidots, (c) dot chains, (d) stripes and (e) dot arrays, grown under different conditions. At elevated temperature, i.e., close to equilibrium growth condition, Co forms quasi-hexagonal three-dimensional (3D) islands (dots). When the nominal thickness is large enough, instead of having the islands to connect into a smooth, continuous film, Co forms a network of flat film with deep quasi-hexagonal holes, i.e., antidots in a film. For both dots and antidots, the edges are along the high-symmetry directions and the surfaces are nearly atomically smooth. The well-defined shape, atomically smooth tops and film network, and the sharp LEED pattern suggest that these islands may be structurally coherent with few defects.

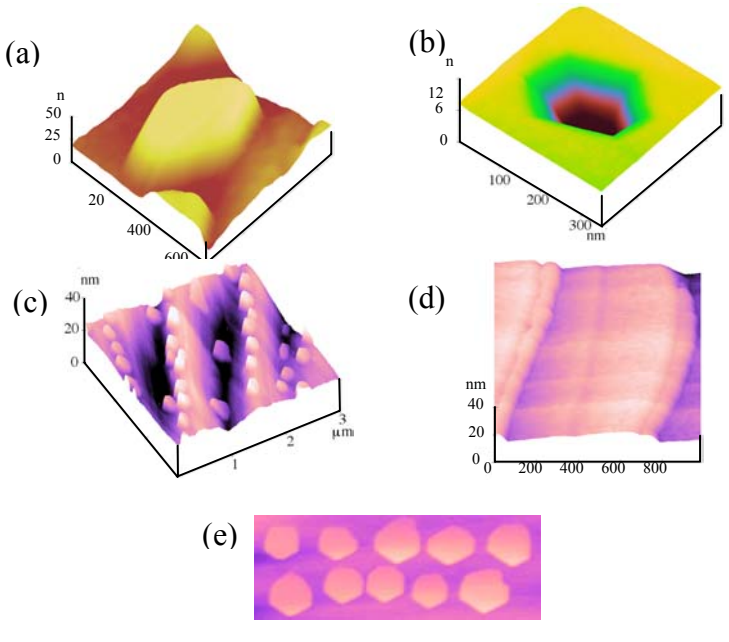

Fig. 1. Typical AFM images of Co (a) dot, (b) antidot, (c) dot chain, (d) stripes, (e) dot arrays.

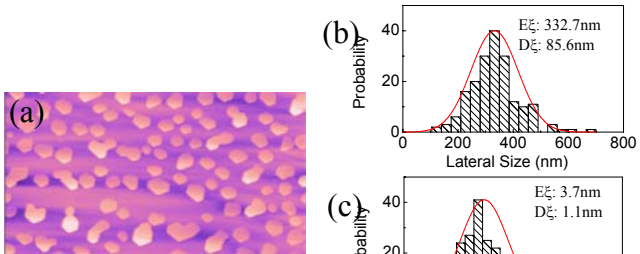

Fig. 2. (a) AFM image of Co dots, and the distribution of (b) lateral size, (c) height, and (d) side-wall angle.

Such a growth is driven by strain relaxation in a highly-ordered epitaxial system. Co has a lower surface energy and a large 8% lattice mismatch with Ru(0001). Instead of forming dislocations in the film, they can form coherent 3D islands to release the strain when growing in S-K mode. As discussed in detail for semiconductor self-assembled quantum dots, such a growth mode prefers uniform dot size since smaller dots grow faster than the large ones, and there exists a minimal size for such dots.[5] And both dots and antidots are equivalent in releasing strain.[18] In Fig. 2, it is shown that the dots have a tendency to be uniform in size and shape, even though the growth conditions are not yet optimized.

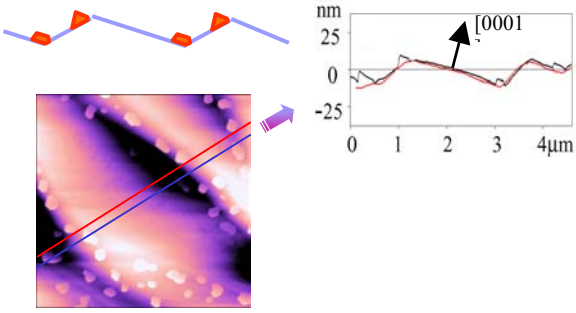

Fig. 3. AFM image of Co dot chains along the grooves on Ru(0001). Line profiles across the grooves, one representing the substrate and the other including Co dots on the grooves, indicate the location of the dots.

While the distribution of the dots on a flat Ru(0001) surface is never completely regular, we have discovered that the placement of these dots can be manipulated with

substrate morphology. The dot chains, stripes and dot arrays in Fig. 1 are formed on grooved Ru substrates, where we left some directional polish scratches. After annealing, the surface reveals a grooved structure with asymmetric saw-tooth profile, as seen in Fig. 3b. The dots align along the top and bottom of the grooved structures into linear chains as in Fig. 1(c). Depending on coverage and groove period, stripes and dot arrays are also observed as seen in Fig. 1(d) & (e).

Figure 3 describes the dot chains in more details. Fig. 3a is a typical AFM image of the self-assembled Co dots on the grooved Ru(0001) at a nominal Co thickness of 1.1 nm. The location of the dot chains is most obvious on Fig. 3b, which shows the line profiles along the two parallel lines across the grooves as indicated on Fig. 3a. Along one line that does not cross any dot, an asymmetric saw-tooth profile of the substrate is apparent. The other parallel line runs across several dots, indicating the location of the dots on the saw-tooth profile. It is clear that the dots grow both at the bottom and the top of the grooves, forming two parallel chains of dots along each edge. More specifically, the ones on top are primarily on the short side of the saw-tooth and the ones at the bottom primarily on the long side. Since the dots are driven by strain relaxation, it is therefore not surprising to see a preferential nucleation of these dots near the top and bottom of the grooves, where strain should be the largest. The fact that the dots sit on only one side of the slope suggests that, besides the strain, other factors such as diffusion along stepped surfaces also play an important role for metals. Noted that the individual dots on grooved substrates show asymmetric profiles perpendicular to the grooves with the top of the dots always in parallel with the long edge (Fig. 3b). We postulate that the growth front of the dots tends to be the stable hcp(0001) plane and therefore result in such an asymmetric shape.

These dots and films with antidots are ferromagnetic. All the dots, ~70-600 nm in diameter, exhibit in-plane single domain states, as seen in Fig. 4a. Our micromagnetic simulations indicate that they are mostly metastable against a vortex ground state, but can easily be trapped in single-domain state depending on history.[19] Indeed, the dots not only have single domain virgin states as observed with MFM, but also exhibit the characteristic square loops for single domains.[15] For thicker films with antidots, the Co magnetocrystalline anisotropy, which is along the c-axis perpendicular to substrate surface, overcomes the in-plane shape anisotropy and forms stripe domains with up-and down perpendicular magnetization (Fig. 4b). Fig. 4c shows that along the dot chains, the dots couple ferromagnetically due to inter-dot magnetostatic interactions, which has been discussed in terms of classical 1D Ising chain. It demonstrates that these self-assembled magnetic dots offer model systems in understanding low dimensional physics.

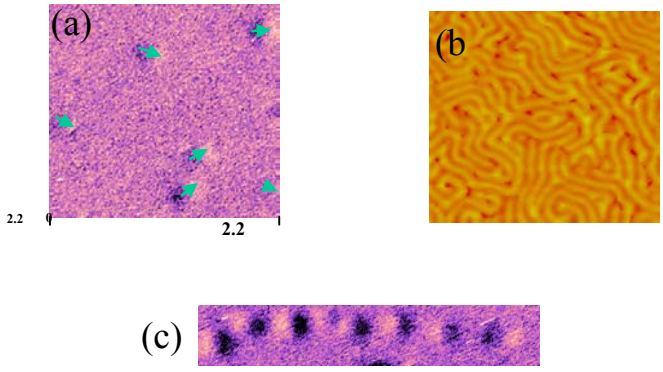

Fig. 4. MFM images for (a) dots, (b) thick film with antidots, and (c) a dot chain.

IV. Summary

We have observe a metal-on-metal growth mode in Co/Ru(0001) at elevated temperature, where self-assembled 3D dots and antidots in rather regular truncated pyramidal shapes are mainly attributed to a stress-driven mechanism. While good lattice match has been one of the major criteria to guide epitaxial growth, it is possible that well-chosen lattice mismatched systems could be utilized to fabricate strain-engineered regular magnetic nanostructure arrays with different sizes and periodicity. A linear alignment of self-assembled Co dots and stripes can be created along grooves on a Ru(0001) substrate. Our observations suggest that it may be possible to direct the alignment and positioning in self-assembly of complex patterns by means of substrate templating, which should be of general applicability beyond the Co/Ru system. Magnetically, the dots are ferromagnetic with in-plane single-domain state, while the thick films with antidots exhibit perpendicular stripe domains.

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